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Arsenic fixation and mobilization in the soils of the Ganges and Meghna floodplains. Impact of pedoenvironmental properties

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ABSTRACT

Arsenic contamination of groundwaters in the floodplain of the Ganges–Meghna river system can represent a serious threat to human health and to the sustainability of irrigated rice cultivation. The extent of As accumulation in the soils irrigated with As-containing groundwater seems to vary among different study areas and the relationships between the pedoenvironmental conditions and As fixation/mobilization are not yet completely elucidated. This work was aimed to assess how soil and environmental properties interact in affecting the fixation/mobilization of As in the soils of the Ganges and Meghna agricultural zones. For this purpose, soil samples from different districts in the Ganges and Meghna floodplains have been characterized, different pools of As, Fe, Mn and P have been quantified and the results have been related to the main soil and environmental characteristics of the two areas. The As content in groundwater and the baseline As concentration in the parent material in the two zones would point to a higher As accumulation in the Meghna floodplain soils, however the Ganges floodplain soils had higher contents of As in all fractions, proving the key role of the factors controlling the release/fixation of As. The soils of the two floodplains, in fact, differed for most physicochemical properties. The ones from the Ganges floodplain were calcareous, with finer texture and generally richer in Fe but oxalate extractable Fe was higher in the Meghna floodplain soils, suggesting a higher degree of waterlogging. This is in agreement with the averagely longer duration and higher depth of submersion of the soils of this area, which enhanced Fe dynamics and favored the release of the less tightly bonded As forms. The competing effect of P was probably similar in the two areas, since P concentrations did not differ significantly among the two soil series. However, more P was Olsen extractable in the Meghna floodplain soils, in contrast with As, that was more easily extracted from the Ganges floodplain soils. The concentration and potential mobility of the retained As were hence greater in the soils of the Ganges floodplain.

1. Introduction

Arsenic (As) contamination of soils and waters is a widespread problem posing a great risk to human health and environmental safety. The contamination of groundwater with As in the Ganges–Brahmaputra–Meghna plain is one of the widest cases of As poisoning in the world (BGS, DPHE, 2001; Brammer and Ravenscroft, 2009; Chakraborti et al., 2004, 2010). In many areas of the Bengal basin, agriculture depends mainly on groundwater for irrigation, and in 40% of the net cultivable area in Bangladesh As contaminated groundwater represents the main water source for irrigation (Huq et al., 2003, 2006). A huge amount of As is thus transferred every year from the contaminated aquifer to the surface water–soil–plant system (Ali et al., 2003a). The As reaching the soil by irrigation could accumulate in the soil solid phase, could be released to the deep or surface waterbodies, could be metabolized and possibly volatilized by microorganisms, and could be taken up by crops from the soil–water system.

Among different crops, rice is particularly subjected to As accumulation because of the required great amounts of irrigation water, potentially As-polluted in this country, and the submerged cropping conditions that enhance the release of the As accumulated in soils to the pore water, which can reach concentrations higher than those of the irrigation water itself (Garnier et al., 2010). In Bangladesh rice cultivation can hence represent one of the main As inputs from groundwater to the soil–crop system (Brammer and Ravenscroft, 2009); moreover, beside rice, also different crops and vegetables grown in As-affected areas, often in rotation with rice, can accumulate high As concentrations (e.g., *arum*), contributing to As dietary intake (Huq et al., 2006; Smith et al., 2006).

The accumulation of As from irrigation water seems to differ largely in Bangladesh agricultural soils (Ali et al., 2003b). In fact, As accumulation could be at least partly counteracted by soil submersion during the monsoon season (Dittmar et al., 2007; Roberts et al., 2010; Saha and Ali,

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2007), but the fraction of the As removed differs among the considered locations (Saha and Ali, 2007) and in several situations a progressive build-up of As content in soils along the years can be foreseen (Dittmar et al., 2010a, 2010b; Khan et al., 2009; Saha and Ali, 2007; van Geen et al., 2006). In other cases, As content in soils does not seem to build up over time, undergoing only seasonal variations around a baseline (Lu et al., 2009). Differences in the pedoenvironment, i.e., soils, climate and other site characteristics including land use, of the studied sites could contribute in explaining these contrasting results, since the overall mobility of the added As depends on the physico-chemical properties of the soils, on As forms, and on the climatic conditions (Begum and Huq, 2007; Huq et al., 2008). In the soil, the quantity and nature of adsorbing phases, mainly iron (Fe) and aluminum (Al) oxides, pH, redox potential, and presence of competing ligands play the most important role (Violante et al., 2011).

Most agricultural soils of Bangladesh develop on the alluvial deposits of the delta formed by the fluvial systems of Brahmaputra, Ganges and Meghna rivers and about 78% are represented by alluvial or flooded soils (FAO, 1988). The soils of the Ganges floodplain, in the south-west of the Country, and of the Meghna floodplain, in the south-east are those subjected to the highest addition of As with groundwater irrigation (Ali et al., 2003a) and differ widely in texture and pH (BARC, www.barc.gov.bd/data_stat.php). Several studies considered potential accumulation and mobility of As in soils in these physiographic units (Hossain et al., 2008; Islam et al., 2004; Rahman et al., 2010; van Geen et al., 2006), but most of the available studies describe As accumulation and/or mobility in one or few sites. Even when soils from different physiographic units are sampled (Saha and Ali, 2007) or very large sample sets are considered (Meharg and Rahman, 2003) the physico-chemical characteristics of the soils are seldom directly related to the differences in the adsorbing phases and to the forms in which As is retained. Thus, the differences in As accumulation in soils are not yet completely explained and a prediction of As fate in the most contaminated sites is still difficult.

The aim of this work was thus to assess how soil and environmental properties interact to affect the amounts of As in the soils of the Ganges and Meghna agricultural areas and to evaluate, through the extraction of As forms, possible differences in the capacity to retain/release As between the soils of the two areas. This would contribute to correctly evaluate the risks linked to the use of As contaminated groundwaters.

2. Materials and methods

2.1. Study areas

The Ganges (GF) and the Meghna (MF) floodplains differ in climatic conditions and soil types. Annual rainfall in GF ranges from 1400 to 2000 mm, but is much higher in MF (2600–2900 mm), and in both cases wide seasonal variations are present (Shahid, 2010). On the average, more than 75% rainfall occurs in the monsoon season, from June to October. The higher amounts of annual rainfall that contribute to flooding and soil submersion (Shahid, 2010), together with local geomorphology induce some variability in land types between the two floodplains. According to the depth of inundation, Bangladesh surface is classified in different inundation land types, from Highlands that are normally not inundated, to Very Lowlands that are inundated above 300 cm depth. The Highlands and Medium Highlands appear to be the most frequent inundation land types in GF, while Medium Lowlands, Lowlands and Very Lowlands are more frequently found in the MF (BARC, www.barc.gov.bd/data_stat.php).

The Bengal Basin is dominated by Holocene alluvial deposits. In the Gangetic plain soils are mostly calcareous and clay–loamy, while in the Meghna floodplain noncalcareous, silty loam to silty clay soils prevail (BARC, www.barc.gov.bd/data_stat.php). The sediment mineralogy is dominated by quartz and feldspars; the clay fraction is composed by illite and kaolinite in nearly equal proportions, whereas chlorite and montmorillonite are in lesser amounts. The sediments of the Ganges system have a higher amount of smectite and a lower amount of kaolinite in comparison to the Meghna ones. In general, the heavy minerals include amphibole, pyrope, and epidote. Some pyrite is also found in the basin sediments (Datta and Subramanian, 1997).

The studied areas (Fig. 1) are located on recent alluvial and deltaic sediments, and belong to the districts of Meherpur, Gopalgaj, Jessore and Sathkhira (GF), Brahmanbaria, Comilla and Chandpur (MF). In agreement with the expected distribution of land inundation types in the two floodplains, in the sampled upazila (i.e., administrative unit within districts, see Supplementary Table), Medium Lowlands, Lowlands and Very Lowlands accounted for 69% of the surface in MF and 22% GF (BARC, www.barc.gov.bd/data_stat.php). According to the British Geological Survey (BGS, DPHE, 2001), the highest average As concentration in groundwater (366 $\mu g\ L^{-1}$) is reported in Chandpur district. The average reported As concentrations for the other districts of the Meghna floodplain studied in this work are 142 $\mu g\ L^{-1}$ for Comilla and 101 $\mu g\ L^{-1}$ for Brahmanbaria district. The average As concentrations for the studied districts of the Ganges floodplain are 187 $\mu g\ L^{-1}$ for Gopalganj, 133 $\mu g\ L^{-1}$ for Satkhira, 116 $\mu g\ L^{-1}$ for Meherpur and 70 $\mu g\ L^{-1}$ for Jessore district. The contamination level of the groundwater in the abovementioned districts, also considering the percentage of contaminated wells, is confirmed by more recent works (Chakraborti et al., 2010).

2.2. Soil sampling, analysis and data treatment

Samples from the topsoil (0–10 cm) were collected in the period June–July 2008 from irrigated paddy fields. Twenty-nine samples were taken in the districts of the Ganges floodplain physiographic unit and 30 in the Meghna floodplain physiographic unit. Rice was generally in rotation with one or more different crops.

The soil samples were air-dried and all the analyses were carried out on the fine-earth fraction (<2 mm). The pH was determined potentiometrically in a 1:2.5 soil/deionized water suspension. The particle size distribution was evaluated by the pipette method after dispersion of the sample with Na-hexametaphosphate; carbonates were determined volumetrically and the C and N contents were determined through dry combustion (NA2100 Protein elemental analyzer, CE Instruments, Milan, Italy). Standard deviations for the above mentioned analysis were within 5% and, for CN determination, within 2%.

The soil samples were analyzed for As, Fe, Mn, P extractable with *aqua regia* (As_R, Fe_R, Mn_R and P_R), with a reducing solution composed by 0.2 M ammonium oxalate–oxalic acid buffered with ascorbic acid (As_{OA}, FeO_{OA}, Mn_{OA} and P_{OA}) shaken at 96 °C for 30 min (Shuman, 1982, as adapted by Wenzel et al., 2001), and with ammonium oxalate buffer in the dark (As_O, Fe_O, Mn_O and P_O Schwertmann, 1964). In this last case Al in the extract was also measured (Al_O). These extractions were aimed to bring in solution different fractions of Fe (Mn and Al) oxides together with the bound As and P, i.e., a pseudo-total fraction, a reducible fraction (mainly Fe and Mn oxides, including well-crystallized Fe oxides), and the poorly ordered fraction of Fe and Al (hydr)oxides. Single-step extractions were adopted instead of sequential extraction procedures in order to avoid some limitations of the sequential extractions, as suggested by Audry et al. (2006), and because of the greater simplicity and rapidity, that appears not to decrease the extraction efficiency (Baig et al., 2009). Bicarbonate extractable As and P (As_{Olsen} and P_{Olsen}, Olsen and Sommers, 1982) were determined as an estimate of the loosely bound pools.

Arsenic was determined by hydride generation (HG) coupled with AAS (Perkin-Elmer 4100 equipped with a FIAS 400 hydride generator; Perkin-Elmer Inc., Waltham, Massachusetts). Iron, Mn and Al were determined with flame-AAS (Perkin-Elmer 3030). Phosphorus was determined colorimetrically (Murphy and Riley, 1962). Standard deviations for all these analysis were within 5%.